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Client Ref. No.: F 04-038-PCT/US/NS

AMENDMENTS TO THE CLAIMS

This listing of claims replaces all prior versions and listings of claims in the application:
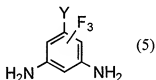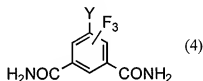
Listing of claims

1. (Previously presented) A method for the production of a fluorinated phenylenediamine represented by the following formula (2), which comprises a step of reacting a diamide represented by the following formula (1) with NaOX [wherein X stands for a bromine atom (Br) or a chlorine atom (Cl)] at a molar ratio of the NaOX to the diamide (NaOX/diamide ratio) in the range of 3.0 - 6.0 and NaOH at a molar ratio of the NaOH to the diamide (NaOH/diamide ratio) in the range of $1.8 - 4.0$,

$$F_l \quad Y_m \qquad (1)$$
$$(CONH_2)_2$$

$$F_l \quad Y_m \qquad (2)$$
$$(NH_2)_2$$
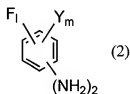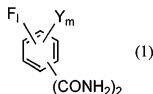
wherein in the formulas (1) and (2), Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C1 - C5 alkyl group optionally having a substituent, or a C1 - C5 alkoxyl group optionally having a substituent, l is an integer in the range of 1 - 4, m is an integer in the range of 0 - 3, provided that the total number of l and m (l + m) is 4; and, upon completion of the reaction, adding an alkali to adjust the pH of the solution to be in the range of 9 to 14.

2. (Original) A method according to claim 1, wherein said diamide is reacted with NaOX and NaOH at a temperature in the range of 0 - 20°C and the resultant reaction product is heated at a temperature exceeding 20°C and not exceeding 100°C.

3. (Previously presented) A method according to claim 1, wherein said diamide is a diamide represented by the following formula (4) and said phenylenediamine is a phenylenediamine represented by the following formula (5),
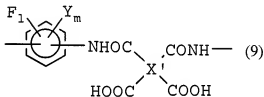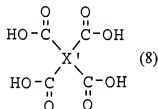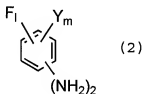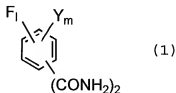

(4)

(5)

wherein in the formulas (4) and (5), Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent.

4. (Previously presented) A method according to claim 1, wherein the molar absorption coefficient of the fluorinated phenylenediamine represented by the formula (2) at a wavelength of 450 nm is not more than 2.5 (l/mol·cm).

5. (Currently Amended) A method for the production of a polyamic acid represented by the formula (9), which comprises the steps of:

producing a fluorinated phenylenediamine represented by the formula (2) by reacting a diamide represented by the formula (1) with NaOX [wherein X stands for a bromine atom (Br) or a chlorine atom (Cl)] at a molar ratio of the NaOX to the diamide (NaOX/diamide ratio) in the range of 3.0 - 6.0 and NaOH at a molar ratio of the NaOH to the diamide (NaOH/diamide ratio) in the range of 1.8 – 4.0, and after the reaction, adding an alkali to adjust the resultant reaction solution pH to be in the range of 9 to 14; and

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reacting the fluorinated phenylenediamine with tetracaraboxylic acid represented by the formula (8), the acid anhydride or acid chloride thereof, or the ester thereof in an organic solvent,

$$F_l \quad Y_m$$
$$(CONH_2)_2 \quad (1)$$

$$F_l \quad Y_m$$
$$(NH_2)_2 \quad (2)$$

$$HO-\overset{O}{\overset{\|}{C}} \quad \overset{O}{\overset{\|}{C}}-OH$$
$$X' \quad (8)$$
$$HO-\overset{}{\underset{\|}{C}} \quad \overset{}{\underset{\|}{C}}-OH$$
$$\overset{}{O} \quad \overset{}{O}$$

$$F_l \quad Y_m$$
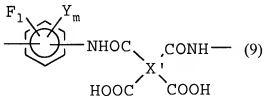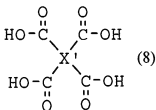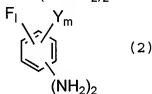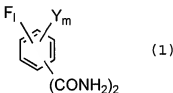$$-NHOC \quad CONH- \quad (9)$$
$$X'$$
$$HOOC \quad COOH$$

wherein Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent, l is an integer in the range of 1 - 4, m is an integer in the range of 0 - 3, provided that the total number of l and m (l + m) is 4, and X' stands for a tetravalent organic group.
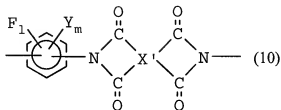
6. (Currently Amended) A method for the production of polyimide represented by the formula (10), which comprises the steps of:

producing a fluorinated phenylenediamine represented by the formula (2) by

reacting a diamide represented by the formula (1) with NaOX [wherein X stands for a
bromine atom (Br) or a chlorine atom (Cl)] at a molar ratio of the NaOX to the diamide
(NaOX/diamide ratio) in the range of 3.0 - 6.0 and NaOH at a molar ratio of the NaOH to
the diamide (NaOH/diamide ratio) in the range of 1.8 – 4.0, and after the reaction, adding
an alkali to adjust the resultant reaction solution pH to be in the range of 9 to 14; and

producing a polyamic acid represented by the formula (9) by reacting the
fluorinated phenylenediamine with tetracaraboxylic acid represented by the formula (8),
the acid anhydride or acid chloride thereof, or the ester thereof in an organic solvent; and
cyclizing by heating the polyamic acid,

$$F_l \diagdown Y_m$$

(1)

$$(CONH_2)_2$$

$$F_l \diagdown Y_m$$

(2)

$$(NH_2)_2$$

$$\underset{HO-C}{\overset{O}{\|}} \quad \underset{C-OH}{\overset{O}{\|}}$$

$$X$$

(8)

$$\underset{HO-C}{\overset{}{\underset{\|}{O}}} \quad \underset{C-OH}{\overset{}{\underset{\|}{O}}}$$

$$F_l \diagdown Y_m$$

$$— \diagdown NHOC \diagdown CONH — \quad (9)$$

$$X$$

$$HOOC \diagup COOH$$

$$\text{(10)}$$

wherein Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent, l is an integer in the range of 1 - 4, m is an integer in the range of 0 - 3, provided that the total number of l and m (l + m) is 4, and X' stands for a tetravalent organic group.

7. (Previously presented) A method according to claim 2, wherein said diamide is a diamide represented by the following formula (4) and said phenylenediamine is a phenylenediamine represented by the following formula (5)

$$\text{(4)}$$

$$\text{(5)}$$
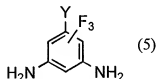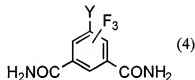
wherein in the formulas (4) and (5), Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent.

8. (Previously presented) A method according to claim 2, wherein the molar absorption coefficient of the fluorinated phenylenediamine represented by the formula (2) at a wavelength of 450 nm is not more than 2.5 (l/mol·cm).

9. (Previously presented) A method according to claim 3, wherein the molar absorption coefficient of the fluorinated phenylenediamine represented by the formula (2) at a wavelength of 450 nm is not more than 2.5 (l/mol·cm).

10. (Previously presented) A method according to claim 7, wherein the molar absorption coefficient of the fluorinated phenylenediamine represented by the formula (2) at a wavelength of 450 nm is not more than 2.5 (l/mol·cm).
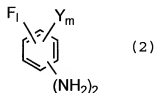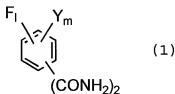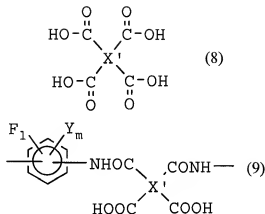
11. (Currently amended) A method for the production of a polyamic acid represented by the formula (9), which comprises the steps of:

producing a fluorinated phenylenediamine represented by the formula (2) by reacting a diamide represented by the formula (1) with NaOX [wherein X stands for a bromine atom (Br) or a chlorine atom (Cl)] at a molar ratio of the NaOX to the diamide (NaOX/diamide ratio) in the range of 3.0 - 6.0 and NaOH at a molar ratio of the NaOH to the diamide (NaOH/diamide ratio) in the range of 1.8 – 4.0, at a temperature in the range of 0 - 20°C, ~~and by~~ heating the resultant reaction product subsequently at a temperature exceeding 20°C and not exceeding 100°C, and, upon completion of the reaction, adding an alkali to adjust the pH of the solution to be in the range of 9 to 14; and

reacting the fluorinated phenylenediamine produced with tetracaraboxylic acid represented by the formula (8), the acid anhydride or acid chloride thereof, or the ester thereof in an organic solvent,
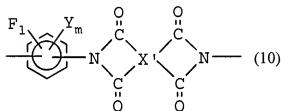
$$F_l \quad Y_m$$

$$(CONH_2)_2 \qquad (1)$$

$$F_l \quad Y_m$$

$$(NH_2)_2 \qquad (2)$$

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$$\begin{array}{c} \text{HO-}\overset{O}{\underset{\|}{C}}\diagdown\overset{O}{\underset{\|}{C}}\text{-OH} \\ X' \\ \text{HO-}\underset{\underset{O}{\|}}{C}\diagup\underset{\underset{O}{\|}}{C}\text{-OH} \end{array} \quad (8)$$

$$F_l \quad Y_m$$
$$\text{—}\bigcirc\text{—NHOC}\diagdown\diagup\text{CONH—} \quad (9)$$
$$X'$$
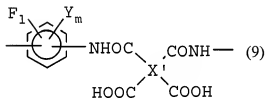$$\text{HOOC}\diagup\diagdown\text{COOH}$$

wherein Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent, l is an integer in the range of 1 - 4, m is an integer in the range of 0 - 3, provided that the total number of l and m (l + m) is 4, and X' stands for a tetravalent organic group.

12. (Currently amended) A method for the production of polyimide represented by the formula (10), which comprises the steps of:

producing a fluorinated phenylenediamine represented by the formula (2) by reacting a diamide represented by the formula (1) with NaOX [wherein X stands for a bromine atom (Br) or a chlorine atom (Cl)] at a molar ratio of the NaOX to the diamide (NaOX/diamide ratio) in the range of 3.0 - 6.0 and NaOH at a molar ratio of the NaOH to the diamide (NaOH/diamide ratio) in the range of 1.8 – 4.0, at a temperature in the range of 0 - 20°C, ~~and by~~ heating the resultant reaction product subsequently at a temperature exceeding 20°C and not exceeding 100°C, and, upon completion of the reaction, adding an alkali to adjust the pH of the solution to be in the range of 9 to 14; and

producing a polyamic acid represented by the formula (9) by reacting the fluorinated phenylenediamine with tetracaraboxylic acid represented by the formula (8), the acid anhydride or acid chloride thereof, or the ester thereof in an organic solvent; and cyclizing by heating the polyamic acid,
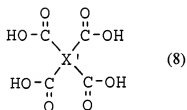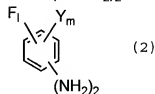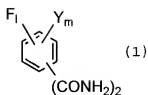
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F_l Y_m (1)

$(CONH_2)_2$

F_l Y_m (2)

$(NH_2)_2$

$HO-C(=O) \quad C(=O)-OH$ — X' — $HO-C(=O) \quad C(=O)-OH$ (8)

F_l Y_m —NHOC   CONH—   (9)

HOOC   X'   COOH

F_l Y_m —N   X'   N—   (10)

wherein Y stands for a hydrogen atom (H), a bromine atom (Br), a chlorine atom (Cl), a fluorine atom (F), a C_1 - C_5 alkyl group optionally having a substituent, or a C_1 - C_5 alkoxyl group optionally having a substituent, l is an integer in the range of 1, m is an integer in the range of 0 - 3, provided that the total number of l and m (l + m) is 4, and X' stands for a tetravalent organic group.

13-15. (Cancelled)

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16. (Previously presented) A method according to claim 1, wherein l is 3 or 4 and m is 0 or 1.

17. (Previously presented) A method according to claim 5, wherein l is 3 or 4 and m is 0 or 1.

18. (Previously presented) A method according to claim 6, wherein l is 3 or 4 and m is 0 or 1.

19. (Previously presented) A method according to claim 11, wherein l is 3 or 4 and m is 0 or 1.

20. ((Previously presented) A method according to claim 12, wherein l is 3 or 4 and m is 0 or 1.